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### (54) BIAXIALLY ORIENTED POLYESTER FILM

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a biaxially oriented polyester film excellent in linear tearability when torn in at least longitudinal direction of the film and suitable as an easily unsealable packaging material.

SOLUTION: This biaxially stretched film is manufactured by stretching lengthwise and crosswise an unstretched film obtained by melt extrusion into a sheet of a raw material prepared by mixing a polybutylene terephthalate (a modified PBT) containing 5-20 wt.% of a polytetramethylene glycol unit having a mol.wt. of 600-4,000 with a polyethylene terephthalate (PET) in a wt. ratio of PET/modified PBT of 70/30-95/5. In this case, a heat of fusion  $\Delta H_c$  (J/g) of the modified PBT and a content X (wt.%) of the modified PBT in the unstretched film satisfy the relation:  $X/200 \leq \Delta H_c \leq X/40$ .

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CLAIMS

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[Claim(s)]

[Claim 1] The polybutylene terephthalate containing 5 - 20 % of the weight of polytetramethylene glycol units of molecular weight 600-4000 (denaturation PBT), The unstretched film obtained by carrying out melting extrusion to the shape of a sheet using the raw material which mixed polyethylene terephthalate (PET) at a rate of PET/denaturation PBT=70 / 30 - 95/5 (weight ratio) It is the biaxially oriented film extended in length and a longitudinal direction. Relation with the content X (% of the weight) of amount \*\*Hc (J/g) of heat of fusions and Denaturation PBT of the denaturation PBT in an unstretched film Biaxial orientation polyester film which is characterized by filling  $X^2 / 200 \leq **Hc \leq X^2 / 40$  and which has \*\*\*\* linearity in the longitudinal direction of a film at least.

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention has the reinforcement of a film which excelled and was excellent in the \*\*\*\* linearity of a longitudinal direction at least, thermal resistance, and dimensional stability, and relates to tearability polyester film useful as wrapping, such as drugs, daily necessities, and cosmetics, including food, such as confectionery, pickles, bean paste, soup, a jam, refrigeration, refrigeration, and a retort pouch.

[0002]

[Description of the Prior Art] The PET film by which many package bags using various kinds of plastic film are used, and biaxial stretching was carried out also in it is excellent in endurance, dampproofing, dynamic reinforcement, thermal resistance, and oilproof, and the sealant film which can be heat sealed, and the package bag laminated more than two-layer are widely used for the package of food, drugs, and miscellaneous goods.

[0003] However, since a PET film has bad tear property, it has the problem that the package bag which carried out the laminating of the sealant film etc. has bad \*\*\*\*\*. Although there is the approach of giving a notch in order to improve opening nature, it not only becomes useless, but the trouble which soft confectionery, such as Cookie, breaks at the time of opening, or soils clothes when contents are liquids may arise by the phenomenon which cannot be torn linearly often occurring, when it tears from a notch, and contents dispersing.

[0004] As an open-easiness ingredient which is excellent in linearity when a film is torn, there are some which laminated the uniaxial-stretching polyolefine film as an interlayer. As such a thing, although there was a three-layer laminate film of biaxial-stretching polyester film / uniaxial-stretching polyolefine film / non-extended polyolefine film, an interlayer must be prepared specially, there is a problem in cost, and the application was limited, for example.

[0005]

[Problem(s) to be Solved by the Invention] This invention tends to solve such a trouble and tends to offer the tearability polyester film useful as wrapping which has the reinforcement of a film which excelled and was excellent in the \*\*\*\* linearity of a longitudinal direction at least, thermal resistance, and dimensional stability.

[0006]

[Means for Solving the Problem] this invention persons reached [ that the above-mentioned technical problem is solved and ] header this invention by adjusting the heat characteristic of an unstretched film in the biaxially oriented film which extended the unstretched film obtained by carrying out the amount combination of specification of the specific polyester resin (denaturation PBT) at PET, and carrying out melting extrusion to the shape of a sheet in length and a longitudinal direction, as a result of inquiring wholeheartedly, in order to solve such a technical problem.

[0007] That is, the summary of this invention is as follows. The polybutylene terephthalate containing 5 - 20 % of the weight of polytetramethylene glycol units of molecular weight 600-4000 (denaturation

PBT), The unstretched film obtained by carrying out melting extrusion to the shape of a sheet using the raw material which mixed polyethylene terephthalate (PET) at a rate of PET/denaturation PBT=70 / 30 - 95/5 (weight ratio) It is the biaxially oriented film extended in length and a longitudinal direction. Relation with the content X (% of the weight) of amount  $H_c$  (J/g) of heat of fusions and Denaturation PBT of the denaturation PBT in an unstretched film Biaxial orientation polyester film which is characterized by filling  $X^2 / 200 \leq H_c \leq X^2 / 40$  and which has  $H_c$  linearity in the longitudinal direction of a film at least.

[0008]

[Embodiment of the Invention] After obtaining oligomer by the well-known process, i.e., the ester exchange reaction method from dimethyl terephthalate and ethylene glycol, or the direct esterifying method from a terephthalic acid and ethylene glycol, PET in this invention can copolymerize other components, if it is a melting polymerization or range which does not spoil the effectiveness of this invention, although solid state polymerization is carried out further and it is obtained.

[0009] As other copolymerization components, oxalic acid, isophthalic acid, a phthalic acid, 2, 6-naphthalene dicarboxylic acid, 5-sodium sulfoisophtharate, a succinic acid, an adipic acid, an azelaic acid, A sebacic acid, dodecane diacid, dimer acid, a maleic anhydride, a maleic acid, Dicarboxylic acid, such as boletic acid, an itaconic acid, a citraconic acid, mesaconic acid, and cyclohexane dicarboxylic acid, Hydroxy acid, such as 4-hydroxybenzoic acid, epsilon-caprolactone, and a lactic acid, 1,3-propanediol, 1,4-butanediol, neopentyl glycol, 1,6-hexanediol, cyclohexane dimethanol, triethylene glycol, Glycols, such as a polyethylene glycol, a polypropylene glycol, and a polytetramethylene glycol, Multifunctional compounds, such as ethylene oxide adducts, such as bisphenol A and Bisphenol S, trimellitic acid, trimesic acid, pyromellitic acid, trimethylol propane, a glycerol, and pentaerythritol, are mentioned.

[0010] the molecular weight of the polytetramethylene glycol (PTMG) used in this invention -- 600-4000 -- desirable -- 1000-3000 -- it is 1000-2000 still more preferably. When  $H_c$  linearity is not acquired at the case of less than 600 but the molecular weight of PTMG exceeds 4000, the  $H_c$  linearity by which engine performance, such as a mechanical strength of a film, dimensional stability, and a haze, was fallen and stabilized is not acquired.

[0011] the ratio of PBT which constitutes Denaturation PBT in this invention, and PTMG -- PBT/PTMG=80 / 20 - 95/5 (weight ratio) -- desirable -- 80 / 20 - 90/10 -- it is 85 / 15 - 90/10 still more preferably. In the  $H_c$  linearity of a film falling since compatibility with PET is too good when PTMG is less than 5 % of the weight, and exceeding 20 % of the weight, about [ that engine performance, such as a mechanical strength of the film obtained, dimensional stability, and a haze, falls ] and film production workability gets worse, thickness spots increase, and  $H_c$  linearity worsens as a result. Moreover, when separating from the range of the above [ the ratio of PTMG under denaturation PBT ], the phenomenon (the so-called ballast phenomenon) which a film ripples at the time of extrusion may occur from the relation of the compatibility of PET and Denaturation PBT. The denaturation PBT in this invention can be acquired also by carrying out melting kneading of PBT and the PTMG with an extruder as a simpler approach, although the polycondensation of the PTMG can be added and carried out and it can be obtained in the polymerization process of PBT.

[0012] the mixed rate of PET and Denaturation PBT in this invention -- PET/denaturation PBT=70 / 30 - 95/5 (weight ratio) -- it is preferably required 80 / 20 - 90/10, and to be referred to as 85 / 15 - 90/10 still more preferably. When the concentration of Denaturation PBT is less than 5 % of the weight, the  $H_c$  linearity of a film is not acquired, but in exceeding 30 % of the weight, engine performance, such as a mechanical strength, dimensional stability, and a haze, falls, a problem arises in practicality ability, or thickness spots become large, and  $H_c$  linearity falls.

[0013] Amount  $H_c$  (J/g) of heat of fusions of the denaturation PBT in the unstretched film in this invention requires that it should ask by carrying out DSC measurement of the unstretched film, and carrying out integral count of the endoergic peak based on fusion of Denaturation PBT, and the content X of the value and Denaturation PBT (% of the weight) should fill the relation between  $X^2 / 200 \leq H_c \leq X^2 / 40$ . In not filling the above-mentioned relation, the  $H_c$  linearity of a film falls. When

**\*\*H** becomes smaller than X2/200, extrusion film production temperature is too high, or when it is easy to become when the residence time is long, and **\*\*H** becomes large from X2/40, kneading in an extrusion film production process is inadequate, or there is an inclination generated when extrusion temperature is too low.

[0014] The residence time says time amount until resin fuses and is extruded from a die through an extruder and single tubing. The residence time will change, if conditions, such as capacity of the rotational frequency of an extruder, the configuration of a screw, single tubing, and a dice and extrusion pressure, change.

[0015] On the film of this invention, other thermoplastics, for example, polyethylenenaphthalate, polycyclohexylene dimethylene terephthalate, etc. are mixable in the range which does not spoil the effectiveness of this invention. Moreover, the organic particle made into a constituent may also contain suitably inorganic particles, such as an ultraviolet ray absorbent, an antioxidant, an antistatic agent, a surface active agent, a pigment, a fluorescent brightener, etc. and a silica, a calcium carbonate, titanium oxide, an acrylic acid, styrene, etc. if needed.

[0016] In order to manufacture the polyester film of this invention, what mixed the chip of Denaturation PBT and PET is supplied to an extruder, after carrying out heating fusion, it extrudes in the shape of a sheet from the die orifice of a T die, and a non-extended sheet is manufactured. It sticks to a cooling drum by the electrostatic impression cast method etc., and twists and cools, next the sheet extruded from the die orifice of a T die is 90-140 degrees C in temperature, is extended by one 2.5 to 5.0 times the scale factor of this in length and a longitudinal direction, respectively, is further heat-treated at the temperature of 210-245 degrees C, and let it be a biaxially oriented film.

[0017] If a homogeneous oriented film may be unable to be obtained and it exceeds 140 degrees C when extension temperature is less than 90 degrees C, crystallization of PET is promoted and transparency may worsen. When draw magnification is less than 2.5 times, the reinforcement of the oriented film obtained is low, and extension may become difficult, if it becomes easy to generate a pinhole and draw magnification exceeds 5.0 times, when it is made a bag. Moreover, if the rate of a heat shrink of the oriented film which will be obtained if heat treatment temperature is lower than 210 degrees C may become large, and the bag after bag manufacture may deform and heat treatment temperature is higher than 245 degrees C, a film may melt.

[0018] As the biaxial-stretching approach, any of a tenter coincidence biaxial-stretching method and the serial biaxial-stretching method by the roll and the tenter are sufficient. Moreover, a biaxially oriented film may be manufactured by tubular **\*\***.

[0019] The surface treatment by corona discharge treatment, surface hardening, plating processing, coloring processing, or various kinds of coating processings can be given to the biaxially oriented film of this invention.

[0020] Next, an example explains this invention concretely. In addition, the measuring method used for evaluation of an example and the example of a comparison is as follows.

[0021] **\*\*\*\*** linearity; from the oriented film of a crosswise center section, 205mm is started to a longitudinal direction (the direction of MD), the piece of a film of the shape of a 20mm strip of paper is started crosswise (the direction of TD), and ten samples which put infeed with a die length of 5mm into the center section of one shorter side of this piece of a film are produced. Next, tore by hand in the direction of MD, and the sample number which reached the shorter side which faces the side into which the **\*\*\*\*** propagation edge put infeed from infeed estimated O and 7-5 as **\*\***, and estimated four or less for eight or more things as x.

[0022] The amount of heat of fusions; DSC measurement was performed about sample 10mg at the programming rate of 20 degrees C / min, and 20-280 degrees C of measuring range using the differential-scanning-calorimetry equipment by PerkinElmer, Inc. From the fusion peak (240-260 degrees C) of PET, integral count was carried out from the fusion peak (200-220 degrees C) of the denaturation PBT by the side of low temperature, and the amount of heat of fusions of Denaturation PBT was calculated.

[0023] Example 1 dimethyl-terephthalate 194 section, 1,4-butanediol 108 section, and tetrabutyl

titanate 80ppm (numeric value converted into the weight of the titanium metal to a polymer) were added, and the ester exchange reaction was performed at 150-210 degrees C for 2.5 hours. After transporting the obtained ester exchange reaction product to the polymerization can and adding 40 ppm of tetrabutyl titanate, PTMG of molecular weight 1100 was added 15% of the weight, reduced pressure was started, finally, under reduced pressure of 4hPa, the melting polymerization was carried out at the temperature of 210-245 degrees C for 2 hours, and Denaturation PBT was acquired. Next, the thing which carried out chip mixing of the acquired denaturation PBT and PET (relative viscosity 1.38) at 15/85 (weight ratio) of a rate 90mmphi extruder possessing a coat hanger type T die is used. Extrusion rotational frequency 42rpm, Adhesion quenching is carried out on the extrusion temperature of 270 degrees C, and the conditions for mean-flow-time 5 minutes, applying the applied voltage of 7kv(s) to melting extrusion and the cast roll by which temperature control was carried out to 20 degrees C at a pinning wire, and it is 210 micrometers in thickness. The unstretched film was obtained. After extending 3.8 times at the temperature of 90 degrees C and extending the obtained unstretched film 4.6 times at the temperature of 120 degrees C by the roll length drawing machine at a tenter tenter, it heat-treated at 230 degrees C, having used the lateral rate of relaxation as 5%, and cooled slowly to the room temperature, and biaxial-stretching polyester film with a thickness of 12 micrometers was obtained. The DSC measurement result of an unstretched film and the evaluation result of the \*\*\*\* linearity of biaxial-stretching polyester film were shown in Table 1.

[0024] Biaxial-stretching polyester film was obtained like the example 1 except having changed the blending ratio of coal of the presentation of the one to examples 2-3 and example of comparison 4 denaturation PBT, and Denaturation PBT and PET, as shown in Table 1. The evaluation result of DSC and \*\*\*\* linearity was shown in Table 1.

[0025] Biaxial-stretching polyester film was obtained like the example 1 except having changed the rotational frequency of five to examples 4-5 and example of comparison 6 extruder, extrusion temperature, and the residence time, as shown in Table 1. The evaluation result of DSC and \*\*\*\* linearity was shown in Table 1.

[0026] Biaxial-stretching polyester film was obtained like the example 1 except having changed the blending ratio of coal of the examples 6-7 and example of comparison 7 denaturation PBT and PET, the rotational frequency of an extruder, extrusion temperature, and the residence time, as shown in Table 1. The evaluation result of DSC and \*\*\*\* linearity was shown in Table 1.

[0027]

[Table 1]

		変性 PBT		配合割合		未延伸フィルムの 押出製膜条件			$X^2 / 200$	$X^2 / 40$	$\Delta H_c$	引裂 直線 性
		P T M G	P B T	変 性 P B T	P B T	押出 機回 転数 rpm	押出 温度 ℃	滞留 時間 分				
実 施 例	1	15	85	15	85	50	270	5	1.1	5.5	2.1	○
	2	5	95	10	90	"	"	"	0.5	2.5	1.1	○
	3	20	80	25	75	"	"	"	3.1	15.5	8.5	○
	4	15	85	15	85	"	262	"	1.1	5.5	3.0	○
	5	"	"	"	"	83	270	3	1.1	5.5	3.5	○
	6	"	"	20	80	"	"	"	2.0	10.0	5.7	○
	7	"	"	10	90	35	"	7	0.5	2.5	0.8	○
比 較 例	1	"	"	3	97	50	"	5	0.1	0.3	0.1	△
	2	"	"	35	65	"	"	"	6.1	30.5	13.0	×
	3	3	97	15	85	"	"	"	1.1	5.5	2.3	×
	4	25	75	"	"	"	"	"	1.1	5.5	2.4	×
	5	15	85	"	"	"	290	"	1.1	5.5	0.6	×
	6	"	"	"	"	25	280	10	1.1	5.5	0.7	×
	7	"	"	10	90	20	255	6	0.5	2.5	2.6	×

[0028]

[Effect of the Invention] According to this invention, it has the reinforcement of a film which excelled and was excellent in the \*\*\*\* linearity of a longitudinal direction at least, thermal resistance, and dimensional stability, and tearability polyester film useful as wrapping, such as drugs, daily necessities, and cosmetics, including food, such as confectionery, pickles, bean paste, soup, a jam, refrigeration, refrigeration, and a retort pouch, is offered.

[Translation done.]